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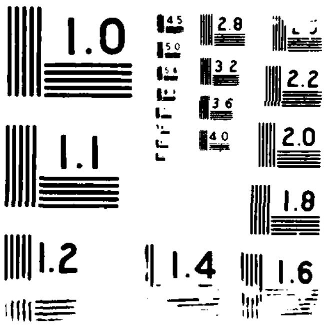
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HIERARCHICAL STRUCTURE IN POLYMERIC SOLIDS AND ITS INFLUENCE ON PROPERTIES

FINAL REPORT

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ABSTRACTS

I. HIERARCHICAL STRUCTURE IN INTERVERTEBRAL DISC AND ITS INFLUENCE ON PROPERTIES

The intervertebral disc is a biological composite whose hierarchical structure is designed to function in a wide range of physiological loading modes. The intervertebral disc is located in the spinal column between the bony vertebral bodies. Their function is to absorb shock and to permit motion between spinal segments. The disc is composed of two parts: a gelatinous nucleus pulposis containing a network of collagen type II fibers, hydrophillic protoglycan molecules, and up to 88% water and the concentric lamellar walls of the annulus fibrosis which is made up primarily of large fibers of collagen type I. The disc is anchored to the vetebral bodies above and below it by cartilage endplates. Optical microscope techniques are used to characterize the hierarchical structure of the collagenous components of the intervetebral disc. These findings are employed in explaining the mechanical response of the disc in compression [1-4].

In the anterior annulus fibrosis, the thickness of lamellae increases abruptly 2 mm inward from the edge of the disc, dividing the annulus into peripheral and transitional regions. Lamellae in the lateral and posterior aspects of the disc have a broad distribution of lamellar thicknesses throughout the annulus. In alternating lamellae, fibers are inclined with respect to the vertical axis of the spine in a layup structure. From the edge of the disc inward to the nucleus, this interlamellar angle decreases from +62 to +45 degrees. Within lamellae, the collagen fibers exhibit a planar crimped morphology. The plane of the waveform is inclined with respect to the vertical axis by the interlamellar angle and fibers are crimped in register. From the edge of the disc inward, the crimp angle increases from 20 to 45 degrees and the crimp period decreases from 26 to 20 um. A hierarchical model of the intervertebral disc has been developed that incorporates these morphological gradients [2].

The significance of this model and the complex gradient structure it represents can be appreciated when examining the mechanical response of the intervertebral disc to compression. Bulging of the disc during compression indicates that axial loads are translated to radial and tangential tensile stresses in the lamellae of the annulus fibrosis. In this case, the expected response of the collagen fibers is reorientation and uncrimping of the waveform as is seen in the tendon and intestine. Such behavior also predicts a compressive stress-strain curve with toe, linear elastic, and yield regions corresponding to the tensile response of the collagen fibers. Compression of an isolated intervertebral disc specimen does show the three-part stress-strain curve characteristic of collagenous tissues in tendon. Preliminary experiments in this laboratory have shown a decrease in crimp angle and increase in crimp period as well as an increase in the interlamellar angle when the disc was fixed under a compressive load. The lamellae will also be most resistant to tensile stress in the directions parallel to the collagen fibers. This agrees with published data on uniaxial tensile testing of specimens cut through several layers of the peripheral annulus that shows maximum stiffness along the two fiber axes [2].

As a result of structural gradients through the annulus, the lamellae are not equally deformable. Peripheral lamellae, with a large interlamellar angle and a small crimp angle, are less deformable than those closer to the nucleus. This is supported by published reports that specimens cut from peripheral lamellae are stiffer and have lower energy dissipation and residual deformation than those of more central lamellae.

II. HIERARCHICAL STRUCTURE IN LIQUID CRYSTALLINE POLYMERS AND ITS INFLUENCE ON PROPERTIES

Thermotropic liquid crystalline polyesters have been described as self-reinforced composites. These materials exhibit similar if not superior mechanical properties to short fiber reinforced thermoplastics. When short glass fibers are added to these liquid crystalline matrices, further enhancement in the mechanical and physical properties are observed. The main objectives in this project were first, to characterize the morphology of these reinforced LCP composites and compared it to the neat LCP resin which is well known to possess several levels of structural organization of different sizes. The second objective was to identify the microfailure mechanisms and fracture behavior of these composites during tensile deformation. Several techniques were applied. For structure characterization, optical microscopy (OM), scanning electron microscopy (SEM) and wide-angle x-ray scattering (WAXS) were employed. As for failure analysis, the approach used was acoustic emission (AE), specifically the number of the acoustic emission signals and their amplitude distributions [5,6].

In the fiber reinforced materials, the optical micrographs taken from different cross-sections parallel and perpendicular to the mold filling direction (MFD), and from in-plane cross-sections at different depths from the surface of the mold, indicated several features to the neat LCP resin. That is, two skin macrolayers in which the glass fibers were found to be mostly oriented parallel to the MFD and a core macrolayer, where the fibers reflected the flow field and were virtually perpendicular to the MFD along the center line of the mold. The corresponding WAXS patterns from the neat LCP resin only at similar depths and locations suggested similar macromolecular orientation. Based on these data, a hierarchical structural model describing these composite materials has been determined. Four types of flow were attributed to such an orientational development of the short glass fibers in the mold. These were: elongational flow, shear flow, converging flow and spreading radial flow. Elongational flow contributed mainly to the high degree of orientation in the surface along the MFD. While converging flow led to some alignment in the MFD, shear flow was predominantly responsible for the orientation at some distance away from the surface. On the other hand, fiber orientation in the transverse direction was attributed to the spreading radial flow.

Typical acoustic emission signals monitored during tensile deformation showed a substantial number of AE events in the composites compared to a few in the neat LCP resin. These few events in the neat resin reflected breakage of the oriented, fribrillar layer at the surface of the mold. High amplitude values above 70-75 dB were their main characteristics. By machining only the top surface, all of the high amplitude events would dissappear.

No high amplitude events above 70-75 dB could be observed in the fiber reinforced composites. The amplitude distribution of the 30 wt% composite was bimodal; with one low maximum at 40-45 dB and a second more predominant maximum between 55 and 70 dB. The low amplitude distribution was associated with breakage of the core macrolayer. From the SEM fracture surfaces, it was concluded that transverse fiber pull-out and matrix cracking in the core were the main microfailure mechanisms which gave rise to these low amplitude values. It was also observed that there was a gradual shift in the amplitude distribution from low to high values. This shift corresponded to growth of parabolic cracks in the core which induced severe damage in the skin macrolayer presumably by stress transfer mechanism, which resulted into the generation of high amplitude events from the skin. Again, from the SEM fracture surfaces, it was concluded that fiber breakage, fiber debonding, fiber pull-out and matrix deformation were the microfailure mechanisms which were responsible for the high amplitude events between 55-70 dB. By a special mini 3-point bending test in the SEM, the sequence of these microfailure processes was identified as following: fiber breakage and small fiber-end cracks, interfacial debonding, localized cracks and large fiber-end cracks, fiber pull-out, interconnection of local and fiber-end cracks leading to catastrophic failure [6].

The 50 wt% composite had only one amplitude maximum at 40-50 dB. Similar cracks in the core macrolayer as the 30 wt% composite were observed. Accordingly, no difference in the amplitude distributions between the core of both composites was seen. This was supported also by the SEM fracture surfaces. The main difference between the two composites was in the skin macrolayer. The absence of the predominant 55-70 dB peak in the 50 wt% composite strongly suggested that some energy absorption mechanisms operating in the skin of the 30 wt% composite no longer operated in the 50 wt%. This has resulted into the premature failure of this composite. SEM fracture surface examination concluded that the premature failure was mainly due to insufficiency in the amount of resin material available to surround effectively the glass fibers. Again, the 50 wt% composite showed similar microfailure mechanisms in the mini 3-point bending test in the SEM, with the exception that less pronounced or almost no matrix deformation was seen.

III. HIERARCHICAL STRUCTURE IN CROSS-ROLLED POLYPROPYLENE AND ITS INFLUENCE ON PROPERTIES

The morphology, mechanical properties and deformation mechanisms of biaxially oriented polypropylene made by hydrostatic solid state extrusion (Bexor process) have been investigated. Structural changes on the spherulitic, lamellar, and macromolecular level were examined by optical, scanning and transmission electron microscopy x-ray diffraction techniques, and differential scanning calorimetry. The results show that polypropylene spherulites undergo stepwise biaxial affine deformation and deform homogeneously into a disc-like morphology. During this spherulitic flattening process, lamellar rotation was found to occur prior to lamellar break-up at a biaxial draw ratio of about 1.5:1. On the macromolecular level, the crystalline c-axis was found to be oriented in the plane concurrently with the lamellar break-up. Amorphous

chains were also oriented preferentially in the plane of deformation. A hierarchical model was proposed to illustrate the nature of orientations in the flattened spherulties. As for the mechanical properties, a wide range of temperatures and strain rates were examined. The anisotropic nature of tensile deformation was analyzed from simultaneous measurements of longitudinal extension and lateral contraction in the width and thickness directions, by determining the shape of the uniaxially deformed spherulites, and by examining the fracture surfaces. It was found that deformation proceeds by elastic extension of the amorphous network and plastic shear deformation of the crystalline regions in the plane of the sheet and in the thickness dimension by voiding with induced fibrillation [7-9].

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